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RESPONSE OF A BURNING PROPELLANT SURFACE TO EROSION TRANSIENTS

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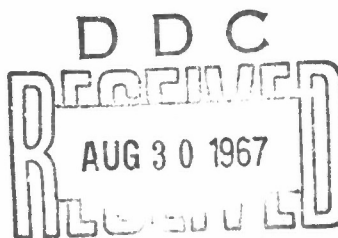
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FOREWORD

The experimental and theoretical studies described in this report are the responsibilities of E. L. Capener and G. A. Marxman, respectively. Major contributors to this research include G. A. Marxman (Principal Investigator), E. L. Capener, C. F. Wooldridge, and R. J. Kier.

The program is under the overall management of the Air Force Office of Scientific Research, Directorate of Engineering Sciences, Propulsion Division (B. T. Wolfson).

NOMENCLATURE

a	frequency factor in Arrhenius law Eq. (2)
C	constant defined by Eq. (7)
c_p	specific heat capacity of gas at constant pressure
c_s	specific heat capacity of solid
E	activation energy for pyrolysis at the interface, Eq. (2)
E_D	activation energy for pressure-insensitive surface-coupled reactions, Eq. (9)
E_f	activation energy for gas-phase reaction, Eq. (7)
E_H	activation energy for pressure-sensitive surface-coupled reactions, Eq. (8)
h	enthalpy
h_{g_w}	= energy carried into gas phase with the vaporizing propellant per unit mass
h_{s_w}	= energy carried by convection from the unreacted solid phase per unit mass
H_D	heat release (positive)/unit mass propellant in pressure-insensitive surface-coupled reactions, Eq. (9)
H_H	heat release (positive)/unit mass propellant (at a reference temperature and pressure) in pressure-sensitive surface-coupled reactions, Eq. (8)
K	thermal diffusivity of solid = $k/\rho_s c_s$
k	thermal conductivity of solid
L	heat of vaporization/unit mass of propellant
m	order of heterogeneous reaction Eq. (8); mass flux from the wall
n	order of gas-phase reaction, Eq. (7)
p	chamber pressure
Q_D	heat of reaction/unit mass of reactant in the pressure-insensitive surface-coupled reaction

Q_H	heat of reaction/unit mass of reactant in the pressure-sensitive surface-coupled reaction
Q_r	heat of reaction/unit mass of reactant in the gas-phase reaction
R	gas constant
r	burning rate
T	temperature
t	time
x	distance into the propellant from its surface

Greek letters

γ	specific heat ratio c_p/c_v
ϵ_r	mass fraction of reactant at the propellant surface (nearly unity)
ρ_s	density of solid propellant
χ	number of sites which undergo surface-coupled reactions per unit mass of solid propellant

Subscripts

f	gas-phase flame
g	gas phase
o	conditions at $x \rightarrow \infty$
s	solid phase
w	conditions at the wall (gas-solid interface)

INTRODUCTION AND SUMMARY

Most of the previous efforts to theoretically determine the response of the solid propellant combustion response to pressure transients have neglected energetic surface reactions altogether. A few earlier analyses that have considered such reactions arrived at conclusions that are at variance with experimental results obtained at SRI and elsewhere. In contrast, the combustion model developed during this investigation is consistent with the observations.

During this quarter, the theoretical effort in this program has focused on developing a clear and complete understanding of the differences between the analyses. This effort is regarded as a prerequisite to proper utilization of the analysis by others and to eventual improvements in the theory during this program. It has been shown that the major difference between the available theories lies in the method of treating surface kinetics. This topic is discussed in detail in this report.

Experimental studies during this quarter have led to the development of a very promising technique for optical studies of the burning propellant surface, using fiber-optics devices. Initial observations based on this technique have provided illuminating information concerning the ammonium-perchlorate deflagration process at the surface and the flame-zone thickness.

THEORETICAL STUDIES

The importance of the combustion response in all unstable combustion situations has been widely recognized, although this investigation, more than others, has emphasized the combustion interaction with traveling waves in the axial mode of the motor chamber. During this investigation our theoretical effort has focused on the formulation of a combustion model that will adequately describe the response of the combustion mechanism to pressure disturbances. This theoretical approach was motivated by two general observations relevant to axial-mode instability in solid-propellant motors: (1) different propellants have widely different stability characteristics in a given combustion chamber,¹ suggesting that the combustion response, rather than gas-dynamical interactions with the chamber and nozzle, is the controlling factor; and (2) those propellants with a relatively high amount of surface-coupled heat release, such as the ammonium perchlorate-based propellants with their significant solid-phase exotherms, are particularly susceptible to unstable combustion.²

Acoustic instability problems have stimulated a number of theoretical studies of the combustion response, which is characterized in terms of an "acoustic admittance," or more generally, in terms of the "response function." Most of these studies, which are comprehensively summarized elsewhere,³ have been basically similar and have neglected surface-coupled reactions entirely. Notable exceptions are the recent analyses by Friedly and Petersen⁴ and by Culick,⁵ in which energetic surface reactions are considered. There have also been several treatments of the effect of surface reactions by Zel'dovich and his colleagues and students in the Soviet Union, recently summarized by Vantoch.⁶ All of these theoretical analyses lead to the conclusion that surface reactions tend to have a stabilizing influence on the propellant. As was noted above, this conclusion appears to be in direct conflict with our experimental observations on composite propellants. Thus, it seems quite

clear that the combustion models underlying these analyses, which differ mainly in mathematical details, do not adequately represent the true combustion mechanism of this class of propellants. Interestingly enough, the analyses appear to describe double-base propellants much better than composites, for the former are observed to have instability regimes that are essentially the opposite of those associated with composites.¹

In an effort to explain this dichotomy, a new combustion model was formulated during this investigation. This model, which differs from earlier ones primarily in the kinetics description of the surface reactions, has been presented in detail previously.² By means of a surface kinetics description that is substantially different from those used in other treatments, the SRI model distinguishes between composite and double-base propellants, unlike other theories. Detailed calculations and comparisons with instability data show that this model is consistent with observations of both classes of propellants. Specifically, surface-coupled exotherms are shown to have a destabilizing effect on composite propellants, whereas pressure-sensitive surface exotherms tend to stabilize double-base propellants. Inasmuch as this result has not been obtained with other treatments, the differences in the analyses are obviously important and deserving of careful consideration.

Theoretical studies during the past quarter have concentrated on developing a comparison of the SRI model⁷ with others. The objective of this comparison was to provide a clear interpretation of the sometimes subtle features that are responsible for the widely different results obtained. Such a comparison is essential for proper interpretation and use of the theoretical results, and also as a basis for our subsequent efforts to improve the model.

Formulation of the Combustion Model

To clarify the differences between the various combustion models, it will be convenient to repeat here the mathematical formulation of the SRI combustion model, as it was presented earlier.^{2,7}

The chief assumptions made in the analysis are the following:

(a) the gas-phase reactions can be represented in terms of a single reaction of arbitrary order that obeys Arrhenius kinetics and responds instantaneously to pressure and temperature disturbances (i.e., time-dependent terms are omitted in the gas-phase equations); (b) the Lewis number is unity in the gas phase; (c) surface pyrolysis and surface-coupled exothermic or endothermic reactions follow Arrhenius laws; and (d) the solid phase is essentially homogeneous with temperature-independent transport properties. For typical propellants, assumption (a) is valid for chamber oscillations at frequencies of a few thousand cps or less. Most of the acoustic instability problems of greatest interest fall within this regime.

As (b) implies, the pyrolysis and surface-coupled reactions are assumed to occur in a surface layer of negligible thickness relative to the penetration depth of the temperature profile. It is difficult to evaluate the quantitative effect of assumptions (b), (c), and (d). However, it is important to remember that all analyses of this kind unavoidably rely on a highly simplified picture of the complex combustion process. Within this context these assumptions are fully justified, even necessary, because they permit a simplified mathematical formulation that is consistent with the underlying concepts.

The formulation begins with the equation governing heat conduction in the solid phase beyond the surface reaction zone:

$$\frac{\partial T}{\partial t} = r(t) \frac{\partial T}{\partial x} + K \frac{\partial^2 T}{\partial x^2} \quad (1)$$

The propellant pyrolysis at the wall is assumed to follow an Arrhenius law so that the burning rate is related to wall temperature as follows:

$$r = ae^{-(E/RT_w)} \quad (2)$$

The following boundary condition is imposed upon the temperature:

$$x \rightarrow \infty; \quad T \rightarrow T_0 \quad (3)$$

The remaining boundary condition is obtained through an energy-flux balance at the gas-solid interface. The net heat conducted into the unreacted solid propellant from the interface at the plane $x = 0$ is

$$-k \left(\frac{\partial T}{\partial x} \right)_w = -k \left(\frac{\partial T}{\partial x} \right)_{g_w} - \rho_s r h_{g_w} + \rho_s r h_{sw} + Q_H + Q_D \quad (4)$$

The first term on the right-hand side of the equality sign represents the energy coming from the gas phase; the second, the energy carried into the gas with the vaporizing propellant; the third, the energy carried by convection from the unreacted solid phase into the interface; the fourth, the energy released (positive) in heterogeneous decomposition reactions at the surface whose reaction rates depend upon the local gas-phase density; and the last, the energy released in solid-phase surface reactions with rates that are independent of gas-phase conditions. It is convenient to rewrite this expression as follows:⁸

$$-k \left(\frac{\partial T}{\partial x} \right)_w = -k \left(\frac{\partial T}{\partial x} \right)_{g_w} + \rho_s r [(c_s - c_p) T_w - L] + Q_H + Q_D \quad (5)$$

Denison and Baum³ have obtained a solution to the gas-phase conservation equations by assuming that the complex gaseous reaction process can be represented by a single-step reaction of order n , where in some cases n may not be an integer. We have retained their gas-phase solution, which yields the following expression for the heat flux from the gas phase to the wall:

$$-k \left(\frac{\partial T}{\partial x} \right)_{g_w} = \rho_s r [\epsilon_r Q_r - c_p (T_f - T_w)] \quad (6)$$

This solution also relates the instantaneous flow of reactant into the gaseous reaction zone, $\rho_s r$, to the instantaneous gas-phase reaction rate so that:

$$r = C_p^{n/2} T_f^{(n/2)+1} e^{-(E_f/2RT_f)} \quad (7)$$

The above derivation assumes that the surface-coupled reactions occur in a thin zone so that the surface heat release acts as a boundary condition on the solid phase. To derive a suitable kinetics description, the solid propellant can be thought of as containing possible reaction sites such that

$$\rho_s r \chi = \begin{array}{l} \text{number of sites which} \\ \text{undergo reaction per} \\ \text{unit area of reaction} \\ \text{zone per unit time} \end{array}$$

where χ is the number of sites that undergo reaction per unit mass of material. The heterogeneous heat release can now be expressed in terms of the above expression and an Arrhenius law as

$$Q_H = \rho_s r H_H \left(\frac{p}{T_w} \right)^m e^{-E_H/RT_w} \quad (8)$$

for a pressure-sensitive reaction. The parameter χ has been absorbed in H_H , the heat release per unit mass. Note that H_H may depend upon the thickness of the surface reaction zone (which is related to $\rho_s r$) and upon the specific character of the pyrolysis process (which is also related ultimately to $\rho_s r$). For example, one might choose to write the above kinetics expression with $(\rho_s r)^y$, instead of $\rho_s r$. Then the exponent y would become an unknown, and somewhat indirect, measure of the extent of surface reactions relative to gas-phase reactions. However, such a modification does not significantly alter the conclusions drawn from the analysis. Therefore, until there emerges a more detailed understanding of the mechanism, any further complications of this type probably are unwarranted and have not been considered.

Except that they are independent of the pressure, the other surface reactions follow a similar law:

$$Q_D = \rho_s r H_D e^{-E_D/RT_w} \quad (9)$$

Equations 5, 6, 8, and 9 can be combined to obtain:

$$-1 \left(\frac{\partial T}{\partial x} \right)_w = \rho_s r \left[\epsilon_r Q_r - L - c_p (T_f - T_o) + c_s (T_w - T_o) + H_H \left(\frac{p}{T_w} \right)^m e^{-(E_H/RT_w)} + H_D e^{-(E_D/RT_w)} \right] \quad (10)$$

Equations 1, 2, and 7, with the boundary conditions of Eqs. 3 and 10, complete the mathematical representation of the combustion model in terms of the dependent variables T_f , T_w , and r . Owing to the nonlinear character of these equations, a closed-form solution cannot be obtained, in general. A linearized solution has been presented in detail previously.²

Comparison of the SRI Theory with Other Response Function Theories

As has been mentioned, the most significant difference between the formulation above and other combustion models lies in the method of accounting for surface reactions. It has been the usual practice in virtually all other theories to employ what may be described as a "one-step" description of surface kinetics. Specifically, the quantities Q_H and Q_D (Eqs. 4 and 5) have been assumed to be proportional to the burning rate r alone. If one follows this procedure, the Arrhenius temperature dependence of the last two terms in Eq. 10 vanishes. This general approach has been used at SRI to describe double-base propellants only, by letting $E_H = E_D = 0$ in Eq. 10. Such a treatment leads to results in good agreement with double-base experiments.² However, it is felt that the more complex "two-step" description of Eq. 10 is necessary for composite propellants, and the experimental results tend to confirm this assumption.² A physical interpretation of the surface reaction process that we are trying to describe in composite and double-base propellants, respectively, will clarify the reasoning that led to the kinetics description incorporated in Eq. 10.

For composite propellants we may envision the surface reaction zone as encompassing, first, pyrolysis of the propellant (described by Eq. 2) and subsequently, surface-coupled reactions among the pyrolysis products (described by Eqs. (8) and (9)). Thus, the activation energies

E_H and E_D associated with Eqs. (8) and (9) represent an overall description of a complicated sequence of events, which may include mixing between the various macroscopic constituents of the composite propellant, as well as reaction steps. In contrast, with a double-base propellant there is no reason to distinguish surface-coupled energetic reactions from pyrolysis reactions, because the constituents are intermixed on a molecular scale. Thus, the pyrolysis and other reactions may be considered as a single sequence, with a single activation energy, E , characterizing the rate-controlling step, and with $E_H = E_D = 0$. A possible exception might occur with heterogeneous reactions, which could call for a very small value of E_H associated with molecular mixing between gas-phase constituents and pyrolysis products.

In summary, according to this interpretation a composite propellant generally corresponds to $E_H > 0$ and/or $E_D > 0$, whereas for a typical double-base propellant $E_H = E_D = 0$. It follows from the SRI theory^{2,7} that exothermic surface-coupled reactions tend to destabilize composite propellants (by increasing the response amplitude), while they tend to stabilize double-base propellants. This rather remarkable theoretical conclusion may explain the previous experimental observation that the stable-burning regimes of composite and double-base propellants are reversed.⁹ It is important to recognize that the possibility of a destabilizing effect associated with surface-coupled exotherms does not arise unless there is a departure from the single-step description of the surface kinetics (i.e., $E_H = E_D = 0$). Because earlier treatments⁴ have employed only the single-step description, they have not predicted this effect.

Relation between Surface Kinetics Description and Steady-State Flame Temperature Behavior Predicted Theoretically

The method of describing surface kinetics in a combustion model is related to the assumed steady-state behavior of the overall heat of combustion, or the flame temperature. It is quite illuminating to consider the surface kinetics question within this context. In steady-state combustion, $-k(\partial T/\partial x)_w = \rho_s r c_s (T_w - T_0)$, in which case Eq. 10

becomes a simple expression for the gas-phase flame temperature in terms of the total heat release in the combustion process. In the present formulation, employing a two-step surface kinetics treatment, the total heat release per unit mass of propellant and the flame temperature tend to rise with increasing pressure because both surface terms increase. (A pressure increase raises r , and therefore T_w .) For a realistic choice of parameters the increase in T_f is small, as it is with real propellants; in fact it can be made equal to that predicted by a thermochemical calculation for any given propellant, if one so desires. This approach, which we have associated with composite propellants, leads to the general conclusion discussed above, i.e., that surface exotherms tend to make the propellant response less stable.

An alternative approach is to retain the two-step kinetics description, but to also require that the total heat of combustion and T_f remain constant. This can be accomplished by imposing the restriction $\theta_s = -(2m/n)(E/RT_w)\theta_H$, from which it follows that the overall pressure exponent $\nu = n/2$. With this restriction the analysis leads to a completely different conclusion; surface exotherms tend to stabilize the propellant. This is the result obtained by others.^{4,5} However, the indicated restriction seems unrealistic. It implies that every exothermic surface reaction will be compensated for exactly by an endothermic reaction, such that the total surface heat release, as well as the overall total (including the gas-phase heat release) will remain constant.

Next we consider the single-step kinetics treatment, in which $E_H = E_D = 0$, so that all surface reactions are characterized by the pyrolysis activation energy. This form of kinetics description is used in all earlier analyses, and it always leads to the conclusion that surface exotherms stabilize the propellant. In the present treatment, where this case is identified with double-base propellants, it is found that the steady-state flame temperature again tends to rise with pressure (unless $m = 0$), as with real propellants. However, if one requires that the flame temperature remain constant ($\nu = n/2$), the present treatment shows that surface reactions have no effect on propellant stability.

It is evident from the foregoing discussion that the method of incorporating surface reactions in the combustion model is a crucial factor. All of the experimental results obtained in our laboratory,^{8,9} associate surface exotherms with instability in composite propellants. Earlier analyses based on simpler kinetics descriptions predict the opposite behavior, whereas the present treatment is consistent with the observations. Additional support comes from the following considerations: (1) The resonance amplitudes and frequencies indicated by the theory are in agreement with experimental observations¹⁰ of typical propellants, within the accuracy of experiment or theory. (2) Observed shifts in the resonance curves and other striking consequences of composition alterations are also anticipated within the context of the theory. (3) In addition, the analysis suggests a plausible explanation for the rather striking differences in the behavior of composite and double-base propellants. These factors strongly suggest that an approach similar to that employed in the SRI analysis is necessary for an acceptable representation of the combustion mechanism.

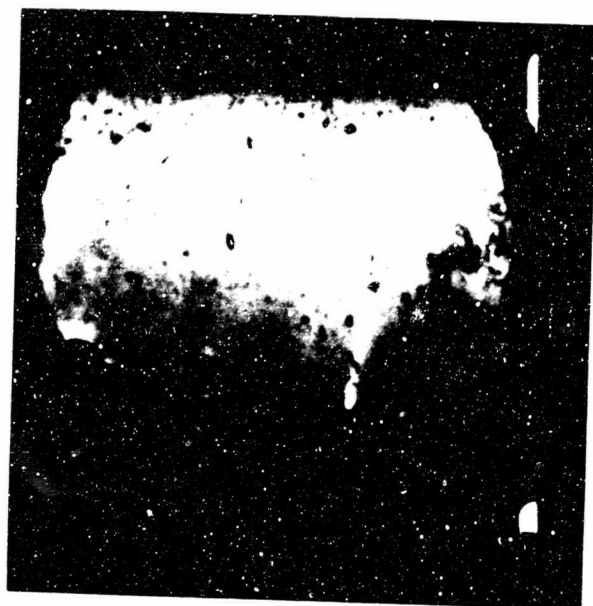
EXPERIMENTAL STUDIES

The preliminary experiments using fiber optics to study the flame zone of burning solid propellants have provided insight into the micro-structure of the flame zone. Magnification in this region is sufficient to reveal details from the surface out to about 2000 microns, a region where the equilibrium flame temperature is reached. Two different experimental techniques are being used in our studies. The first involves using a single 1/8-in. dia. fiber optic embedded in a 1/2-in. thick x 1-5/8-in. dia. propellant pellet which is burned as an end-burning grain. The combustor is 2-in. I.D. x 6-in. long and is terminated by the sample holder at one end and a copper nozzle at the other end. During combustion the image at the end of the fiber optic is photographed at framing rates varying from 1000 to 8000 fps. The second technique for studying the flame zone is by shadowgraph photography using a mercury lamp for illumination. This requires a second fiber optic to back-light the combustion zone.

The initial photographs without back lighting have revealed that the combustion zone is frequently disturbed by the explosive deflagration of single ammonium perchlorate (AP) crystals. This phenomenon is observed more clearly when camera framing rates on the order of 1000 fps are used. At a combustion pressure of 200 psig during the burning of an 80/20 AP-polyurethane propellant, the deflagration of single crystals appears in less than 1/2% of the AP present. This estimation of frequency was made on the basis of a particle count of the deflagration and the estimated depth of field of the optical setup. Several photographs of these deflagrating crystals are reproduced in Fig. 1.

The early shadowgraph pictures of the flame zone have revealed that the equilibrium flame temperature of the combustion gases from an 80/20 AP-polyurethane binder propellant burning at 200 psig is attained at a distance of 1200-1500 microns from the burning surface. This has been inferred from the film density gradient occurring across the combustion zone. We are attempting to correlate the film image density fluctuation

with the actual gas density changes taking place during transit through the combustion zone. Our conclusions on the flame zone thickness are an order of magnitude greater than the early data reported by Summerfield et al.¹¹ However, more recent measurements by Tourin et al.,¹² Povinelli,¹³ and Waesche¹⁴ using both infrared and visible light spectrographic techniques have given values in the range from 1000 to 2000 microns, which is in agreement with our measurements.



(a)



(b)

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FIG. 1a AND 1b DEFLAGRATION OF SINGLE
AMMONIUM PERCHLORATE
CRYSTALS — COMBUSTION
PRESSURE 200 psig —
1 cm = 440 MICRONS

FUTURE STUDIES

The chief purpose of the linear analysis discussed above and previously² has been to provide a preliminary comparison with our experimental data as a check on the hypotheses implied by the combustion model. As has been explained, the comparison strongly supports the theoretical approach. Consequently, we are now undertaking a nonlinear analysis of the combustion model. The nonlinear treatment, which is obtained by programming Eqs. (1), (2), (3), (7), and (10) for a digital computer, will permit us to analyze the combustion response to finite-amplitude waves of any wave form. This program will be used for a more detailed and more quantitative correlation of our data than is possible with a linear analysis. The development of this computer program is in progress, and the first effort at correlation of data will begin during the coming quarter.

The fiber-optic technique for examining burning propellant surfaces, which was developed as part of this investigation, shows great promise as an experimental tool. Already it has been possible to make important observations concerning AP deflagration and the combustion zone thickness (see discussion above). Future experimental studies will concentrate on further development of this technique for use in examining the burning surface of an actual rocket motor. Ultimately, we plan to conduct optical studies of the combustion process as traveling waves, induced by the pulse technique used in this study,^{1,9} pass over the surface. These experiments should provide significant new information concerning transient combustion processes in solid propellant motors.

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